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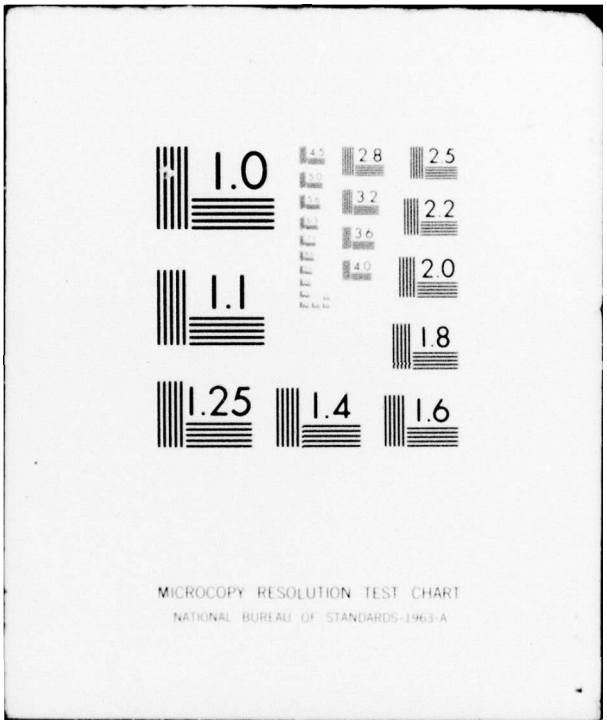
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Gas surface interactions have been studied in the first year of operation of a new state-of-the-art apparatus that can study individual gas-surface collisional events or the velocity distributions of recombining species. Systems studied included Tungsten-O_2, N_2, H_2, He, Ar and Platinum-He, O_2/CO, O_2/C_2H_2, O_2/C_2H_4. The direct reaction of oxygen with tungsten has been characterized. The indirect molecular precursor reaction of N_2 with tungsten is under investigation. The highly non-thermal CO_2 produced by oxidation of CO, C_2H_2 or C_2H_4 on Pt has been clearly established.

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Progress to date -- Gas Surface Interactions
March 1, 1977 - January 1, 1978

Personnel:

The following graduate students have participated in the surface dynamics research activities described below:

Charles Becker, James Cowin, Jerry Hurst, Jihwa Lee.

Dr. Ken Janda joined in our activities in August, 1977, as a visiting NSF postdoctoral fellow. Professor Daniel Auerbach from Johns Hopkins University continues to offer invaluable consulting help.

Apparatus-Instrumentation

The surface apparatus continues to function with a high degree of availability. We acquired a floppy disk for our pdp-11 computer (AFOSR funds) in order to store, manipulate and facilitate transfer of the extension files of surface scattering data. We are performing interpretive and model-fitting calculations on our pdp-11 and the University's IBM 370/168. Currently we are switching from BASIC to FORTRAN operating systems for on line control of experimentaion.

We have constructed a gas recirculating system that recycles valuable gases and gas mixtures used for the supersonic source. This enables us to use enriched isotopes such as $^{15}N - ^{15}N$ and expensive rare gases such as xenon with little actual consumption.

As we shake down the apparatus in the first year of operation we have eliminated troublesome bugs. The most vexing bug was a spurious noise that crept into the signal when high background counting rates were present. It was tracked down and eliminated by performing an autocorrelation computation on the background time-of-flight spectrum. This spectrum proved to have a non-random time autocorrelation. By finding how this phenomenon varied with differing operating conditions, it was tracked down to spurious pickup of rb radiation from the quadrupole mass filter that triggered the time-of-flight initiation pulse.

The principal investigator crashed the detector into one of the main chamber walls. This has bent the detector viewing axis away from its center of rotation by about 0.5 mm. While this defect is not fatal, plans have been drawn up to bend the detector axis back to the center of rotation at the next scheduled major shutdown.

The productivity of the apparatus is enormous in terms of acquisition of data: we have filled ten thick loose leaf notebooks with printouts and plots of surface scattering information. Because the apparatus runs reliably under computer control, it is routinely run overnight and over weekends. By this means enough signal can be acquired from experiments that yield low data rates.

Scientific Results

A useful means of presenting our activities is a listing of systems studied:

Tungsten - Oxygen
Nitrogen
Hydrogen
Helium
Argon

Platinum - Helium
Oxygen/Carbon Monoxide
Oxygen/Acetylene
Oxygen/Ethylene.

Some of the activities of the year have been exploratory studies of various systems as a means of selecting phenomena for deeper study. Other work has investigated specific systems in depth.

I will comment on those areas where the greatest scientific significance lies:

Tungsten-Oxygen. We have shown by examining the O_2 scattered from clean reactive tungsten surfaces that the small fraction of unreacted O_2 observed is essentially elastically scattered. On clean tungsten the reaction probability is high and independent of surface temperature from about 1000-3000° K. At about 400° K surface temperature the sticking coefficient is somewhat higher. The dynamical evidence shows that at the higher surface temperatures there is a fast direct chemical reaction. At the lowest surface temperatures, an additional reaction channel appears. It is probably due to a molecular precursor state. On unreactive oxidized tungsten surfaces, there is large angle diffusely appearing scattering of O_2 (and He) which has been shown to be essentially elastically scattered.

Tungsten-Nitrogen. Prior work has established this system as dominated by a classical molecular precursor reaction. But a controversy exists as to whether the observed decline in sticking coefficient of N_2 (to form a chemisorbed surface atomic nitrogen state) is due to a drop in the condensation coefficient of N_2 on hot W, or due to an increase with surface temperature of the re-evaporation rate of N_2 previously condensed. We have performed extensive studies of this system. We used ^{15}N-^{15}N to permit observation of the time-of-flight of scattered N_2 at the 30 AMU setting of our mass spectrometer where there is relatively low background compared to 28 AMU. The data, which are currently under scrutiny, lead to the conclusion that neither one or the other interpretation can be fully correct, and that both processes lead to a decline in sticking coefficient with increasing surface temperature. The relative importance of the two phenomena depends upon incident gas velocity and surface temperature. A clearer description of the process of condensation of a molecule on a surface will emerge from our studies of this prototypical molecular precursor surface reaction

Tungsten-hydrogen, helium, Neon, Argon. We have studied angular and velocity distributions of scattered H_2, He, and Ar from poly-crystalline tungsten. We searched for a two-dimensional diffraction pattern that would be analogous to a 3-dimensional x-ray powder pattern. Instead we observed with He a multiplicity of zeroth order diffraction peaks, each one sharp and distinct from each other. A significant observation showed that

the He diffraction peak intensities were affected by presence of hydrogen on the surface. As the surface concentration of chemisorbed hydrogen increased from zero, the He diffraction peaks initially diminished in amplitude. As the hydrogen coverage approached saturation (equivalent dosing pressure $\sim 10^{-6}$ torr) the peaks reappeared and were stronger than for the clean surface.

We have searched--without success--for direct evidence of collisional excitation of W-H and W-O vibrational transitions by observation of inelastic scattering of He, Ne, and Ar. Apparently the effect is too small to be observed with our present apparatus. We can however characterize the degree of elasticity of collisions of He and Ne on clean and oxidized W. We are currently designing a high performance nozzle system to investigate this phenomenon more fully.

We have constructed and made preliminary performance tests in a test chamber of an electron gun for observation of electron-desorbed neutrals.

Publications and Manuscripts

It is evident that much of our work is in the "incubation" state. Work from a new apparatus of this sort requires considerable testing and review before it is fit for public scrutiny. Systematic errors are uncovered by critical evaluation and comparison of a range of data from a range of operating conditions. In our first year of operation we have learned that by relatively simple changes in hardware or operating conditions much greater information may be obtained about a given system.

Publications

1. Energy Accommodation and Reactivity of O_2 on Tungsten, by Daniel Auerbach, Charles Becker, James Cowin and Lennard Wharton. Appl. Phys. 14, 141 (1977).

2. Mechanism and Speed of Initial Step of Oxygen Chemisorption - O_2 on W, by Daniel Auerbach, Charles Becker, James Cowin and Lennard Wharton. Appl. Phys. 14, 411 (1977).

3. CO_2 Product Velocity Distributions for CO Oxidation on Platinum, by Daniel Auerbach, Charles Becker, James Cowin and Lennard Wharton. J. Chem. Phys. 67, 3394 (1977).

4. Absence of Translational Energy Accomodation of O_2 on Clean and Oxidized Tungsten, Specularly and Diffusely Scattered, by Daniel Auerbach, Charles Becker, James Cowin and Lennard Wharton. Book of Abstracts (VI International Symposium on Molecular Beams, Noordwijkerhout, The Netherlands, 1977) Part II, p. 192.